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## (54) PRODUCTION OF CORDIERITE CERAMIC BODY

## (57)Abstract:

PROBLEM TO BE SOLVED: To provide a production method of a cordierite ceramic body having a coefficient of thermal expansion similar to the one obtained by using the original starting raw material without necessitating strict control of pulverizing conditions while keeping a good yield of a regenerated raw material when producing the cordierite ceramic body using the unburned regenerated raw material recovered in the production process of the cordierite ceramic body.

SOLUTION: At least a part of a binder is removed from the regenerated raw material which is recovered in the production process of the cordierite ceramic body and comprises a prescribed formulation for cordierite raw material. The cordierite ceramic body is produced by pulverizing the regenerated raw material thus obtained to prepare pulverized powder, adding water, a binder, etc., to the pulverized powder, kneading it to prepare regenerated body, compacting the prepared body and firing it.

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(54) 【発明の名称】 コージェライト・セラミック体の製造方法

(57) 【要約】

【課題】 コージェライト・セラミック体の製造過程において回収される未焼成の再生原料を用いてコージェライト・セラミック体を製造する際に、厳密な粉碎条件制御が不要で、再生原料の歩留まりが良く、しかも元来の出発原料を用いた場合と同程度の熱膨張係数を示すコージェライト・セラミック体の製造方法を提供する。

【解決手段】 コージェライト・セラミック体の製造過程において回収される、所定のコージェライト化原料調合物からなる再生原料中に含まれるバインダーの少なくとも一部を再生原料から取り除いた後に、該再生原料を粉碎して粉碎粉を作製し、該粉碎粉に水分、バインダー等を加え、かつ混練することによって再生坯土を作製し、該再生坯土を成形し焼成してコージェライト・セラミック体を製造する。

## 【特許請求の範囲】

【請求項1】 コージェライト・セラミック体の製造過程において回収される、所定のコージェライト化原料調合物からなる未焼成の再生原料からコージェライト・セラミック体を製造する方法であって、再生原料中に含まれるバインダーの少なくとも一部を再生原料から取り除いた後に、該再生原料を粉碎して粉碎粉を作製し、該粉碎粉に水分、バインダー等を加え、かつ混練することによって再生坯土を作製し、該再生坯土を成形し焼成することを特徴とするコージェライト・セラミック体の製造方法。

【請求項2】 粉碎粉に元来の出発原料及び、水分、バインダー等を加え、かつ混練することによって再生坯土を作製することを特徴とする請求項1に記載したコージェライト・セラミック体の製造方法。

【請求項3】 加熱することにより、再生原料中に含まれるバインダーの少なくとも一部を再生原料から取り除くことを特徴とする請求項1に記載したコージェライト・セラミック体の製造方法。

【請求項4】 バインダーとしてメチルセルロースを使用することを特徴とする請求項3に記載したコージェライト・セラミック体の製造方法。

【請求項5】 280℃以上の温度で加熱することにより再生原料中に含まれるバインダーの少なくとも一部を再生原料から取り除くことを特徴とする請求項4に記載したコージェライト・セラミック体の製造方法。

【請求項6】 280℃以上580℃以下の温度で加熱することにより再生原料中に含まれるバインダーの少なくとも一部を再生原料から取り除くことを特徴とする請求項4に記載したコージェライト・セラミック体の製造方法。

## 【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明はコージェライト・セラミック体の製造過程において回収される未焼成の再生原料を用いてコージェライト・セラミック体を製造する、コージェライト・セラミック体の製造方法に関する。

【0002】

【従来の技術】コージェライト・セラミック体は耐熱性を有し、また広い温度範囲において、低い熱膨張係数を示すことが知られている。このためコージェライト・セラミック体は、高い耐熱衝撃性が要求される排気ガス浄化用ハニカム触媒担体として特に注目されている。通常、コージェライト・セラミック体を製造する場合は、カオリン、タルク、アルミナ等のセラミック原料に溶媒、有機バインダー等の成形助剤を混合、混練して得た坯土をハニカム構造体用の口金を通して押出成形した後、焼成する。コージェライト・セラミック体の熱膨張係数を低くするためには、上記製造工程において、出発

原料の粒子の直径、原料組成等を最適化する必要がある。コージェライト・セラミック体を経済的に製造するには、成形工程から焼成工程に移行する際に除外される未焼成の乾燥成形体またはその破片等の廃棄物を再生原料として再生使用することが望ましい。

【0003】

【発明が解決しようとする課題】しかしながら、再生原料として使用するために未焼成の乾燥成形体またはその破片等の廃棄物を粉碎すると、その粉碎粉を用いて製造したコージェライト・セラミック体は多くの場合、元来の出発原料を用いて製造されたコージェライト・セラミック体に比べて熱膨張係数が大きく耐熱衝撃性が低下して、排気ガス浄化用ハニカム触媒担体として使用できなくなるという問題がある。そこで、そのような問題の対処方法として、コージェライト・セラミック体の加圧成形面におけるプロトエンスタイト面とコージェライト面とのX線回折ピーク強度が一定値になるように、再生原料を調整する方法が提案されている（特公平3-72032）。この特許中では再生原料を粉碎する具体的な方法として再生原料を歯付きロールクラッシャーで粗粉碎後、ピンミルを用いて微粉碎することが望ましいとの記載がある。しかしながらピンミル粉碎におけるピンの本数、周速、材料の投入量により、再生原料に差が生じ、得られたコージェライト・セラミック体の熱膨張係数は大きく変化し、それらの粉碎条件を厳密に制御する必要がある。また最適な条件で粉碎を行った場合でも、元来の出発原料を用いて製造されたコージェライト・セラミック体に比べて $0.6 \times 10^{-7} / ^\circ\text{C}$ ほど高い熱膨張係数を示し、同等のものは得られていない。また厳密な粉碎条件制御が不要な方法として、粒径1mm未満の粉碎粉を除去し、残った粉碎粉のみ使用する方法が提案されている（特開平8-119726）。しかしながらこの方法は粒径1mm未満の粉碎粉を除去するため歩留まりが悪く経済的に問題がある。本発明はコージェライト・セラミック体の製造過程において回収される未焼成の再生原料を用いてコージェライト・セラミック体を製造する際に、厳密な粉碎条件制御が不要で、歩留まりが良く、しかも元来の出発原料を用いた場合と同程度の熱膨張係数を示すコージェライト・セラミック体の製造方法を提供することを目的とする。

【0004】

【課題を解決するための手段】本発明はコージェライト・セラミック体の製造過程において回収される、所定のコージェライト化原料調合物からなる未焼成の再生原料からコージェライト・セラミック体を製造する方法であって、再生原料中に含まれるバインダーの少なくとも一部を再生原料から取り除いた後に、該再生原料を粉碎して粉碎粉を作製し、該粉碎粉に水分、バインダー等を加え、かつ混練することによって再生坯土を作製し、該再生坯土を成形し焼成することを特徴とするコージェライ

ト・セラミック体の製造方法である。本発明において最も重要な点は、上記再生原料からバインダーの少なくとも一部を取り除くことである。

【0005】

【作用】再生原料を使用する際には、水分、バインダー等との均一な混合が容易になるように、混練する前に粉碎して適切な大きさに分解することが望ましい。しかし再生原料はメチルセルロース、PVA、CMC、澱粉のり、グリセリン等のバインダーで比較的強固に結合されているため、粉碎には比較的大きな力が必要となる。ところが、コーゼライト化原料として通常用いられるタルク、カオリン等の層状形状を有する原料粒子は比較的柔らかく、機械的衝撃に対して壊れやすい。特公平3-72032においては、粉碎時の衝撃により原料粒子に微細なクラック、更には結晶構造のメカノケミカル的反応が惹起されやすく、このためにコーゼライト反応過程で元来の出発原料を使用した場合とは全く異なった反応を起こすため、熱膨張係数が上昇するという記載がある。そこで粉碎前に再生原料からバインダーの少なくとも一部を除去することによりバインダーの結合力を低減

【0006】

【発明の実施の形態】本発明の実施例におけるコーゼライト・セラミック体の製造方法について説明する。使用する再生原料は表1のような配合のセル壁厚さ0.17mm、ピッチ1.27mm、長径143mm、短径98mm、長さ152.4mmのハニカム状の未焼成の乾燥成形体である。この成形体2kgに対して昇温速度7.5℃/分にて280℃または580℃まで昇温後、炉冷した。室温まで冷却後、ロールクラッシャにて成形体を粉碎して粉碎粉を作製した。この時、粉碎をハニカム構造を崩すのみに留める為、ロール面間距離は長径方向のハニカム壁の厚さの合計よりわずかに大きい20mmとした。その粉碎粉100部に対して、水31.5

部、メチルセルロース3.5部、ステアリン酸0.5部を加え、混練して再生坯土を作製した。この再生坯土を金型を用いた押出成形法により成形した。この成形体はセル壁厚さ0.17mm、ピッチ1.27mm、直径30mm、長さ100mmのハニカム体である。次にこの成形体を高周波乾燥機を用いて乾燥した後、1400℃、4時間焼成してハニカム状のコーゼライト・セラミック体を得た。

【0007】

【表1】

| 成分      |          | 配合 (wt %) |
|---------|----------|-----------|
| セラミック原料 | 生カオリン    | 20.0      |
|         | 仮焼カオリン   | 10.0      |
|         | タルク      | 40.0      |
|         | アルミナ     | 15.0      |
|         | 水酸化アルミ   | 7.0       |
| 有機バインダー | シリカ      | 8.0       |
|         | メチルセルロース | 3.5       |
| 潤滑剤     | ステアリン酸   | 0.5       |
| 水       |          | 31.5      |

【0008】比較例として、以下の方法でコーゼライト・セラミック体を製造した。表1の乾燥成形体をインバクトクラッシャを用いて粉碎した。ここでインバクトクラッシャとは、固定されている衝突板と、回転するローターに取り付けられた打撃板の間で粉碎を行う装置である。粉碎後、粉碎粉を各種の篩を用いて、0.2mm未満、0.2mm以上0.5mm未満、0.5mm以上1.0mm未満、1.0mm以上2.0mm未満、2.0mm以上4.0mm未満の5種類に選別した。それぞれの粒径範囲の粉碎粉100部に対して、水30.3部を加え、混練して再生坯土を作製した。この再生坯土を金型を用いた押出成形法により成形した。この成形体はセル壁厚さ0.17mm、ピッチ1.27mm、直径30mm、長さ100mmのハニカム体である。次にこの成形体を高周波乾燥機を用いて乾燥した後、1400℃、4時間焼成してハニカム状のコーゼライト・セラミック体を得た。また元来の出発原料を使用して、表1の乾燥成形体と同じ配合の坯土を作製し、上記と同じ方法で成形、焼成してハニカム状のコーゼライト・セラミック体を得た。

【0009】実施例、比較例で作製したコーゼライト・セラミック体の40～800℃における平均熱膨張係数を測定した。結果を表2に示す。実施例においては加熱温度が280℃、580℃のいずれにおいても、元来の出発原料を使用したコーゼライト・セラミック体と同程度の熱膨張係数が得られた。インバクトクラッシャにより粉碎した粉碎粉においては、粒径0.5mm未満では粒径の低下と共に熱膨張係数が上昇し、粒径0.5mm以上では熱膨張係数はほぼ一定となっている。しかしながらいずれの粒径における熱膨張係数も元来の出発

原料を使用したコーゼライト・セラミック体に比べて \*【0010】  
高くなっている。 \* 【表2】

|          | N<br>o. | 再生原料の粉砕方法                 | 粒径範囲 (m)       | 熱膨張係数<br>( $\times 10^{-7}$<br>/℃) |
|----------|---------|---------------------------|----------------|------------------------------------|
| 実施<br>例  | 1       | 280℃に加熱後、ロールク<br>ラッシャにて粉砕 |                | 6.3                                |
|          | 2       | 580℃に加熱後、ロールク<br>ラッシャにて粉砕 |                | 6.2                                |
| 比 較<br>例 | 3       | インパクトクラッシャ<br>により粉砕       | 0.2未満          | 12.2                               |
|          | 4       |                           | 0.2以上0.5<br>未満 | 9.6                                |
|          | 5       |                           | 0.5以上1.0<br>未満 | 8.5                                |
|          | 6       |                           | 1.0以上2.0<br>未満 | 8.5                                |
|          | 7       |                           | 2.0以上4.0<br>未満 | 8.6                                |

【0011】実施例No. 1の再生原料と元来の出発原料を下記表3に示される割合で配合し、更にこの配合物100部に対して、水31.5部、メチルセルロース3.5部、ステアリン酸0.5部を加え、混練して再生坯土を作製した。また実施例1における比較例No. 7の再生原料と元来の出発原料を下記表3に示される割合で配合し、適量の水、メチルセルロース、ステアリン酸20を加え、混練して再生坯土を作製した。これらの再生坯土を金型を用いた押出成形法により成形した。この成形体はセル壁厚さ0.17mm、ピッチ1.27mm、直径30mm、長さ100mmのハニカム体である。次にこの成形体を高周波乾燥機を用いて乾燥した後、1400℃、4時間焼成してハニカム状のコーゼライト・セ※

※ラミック体を得た。

【0012】上記で作製したコーゼライト・セラミック体の40～800℃における平均熱膨張係数を測定した。結果を表3に併せて示す。実施例においては、どのような割合で配合しても、元来の出発原料を使用したコーゼライト・セラミック体と同程度の熱膨張係数が得られた。これに対して、比較例においては、配合比率10%までは元来の出発原料を使用したコーゼライト・セラミック体と同程度の熱膨張係数が得られるが、それ以上配合すると、熱膨張係数の上昇が認められる。

【0013】

【表3】

| N<br>o. | 使用再生原料 | 配合比率            |         | 熱膨張係数<br>( $\times 10^{-7}$<br>/℃) |
|---------|--------|-----------------|---------|------------------------------------|
|         |        | 元 来 の 出 発<br>原料 | 再 生 原 料 |                                    |
| 8       | —      | 100             | 0       | 6.2                                |
| 9       | No. 1  | 80              | 20      | 6.2                                |
| 10      | No. 1  | 60              | 40      | 6.3                                |
| 11      | No. 1  | 40              | 60      | 6.2                                |
| 12      | No. 1  | 20              | 80      | 6.2                                |
| 1       | No. 1  | 0               | 100     | 6.3                                |
| 13      | No. 7  | 95              | 5       | 6.3                                |
| 14      | No. 7  | 90              | 10      | 6.2                                |
| 15      | No. 7  | 80              | 20      | 7.4                                |
| 16      | No. 7  | 60              | 40      | 8.7                                |
| 7       | No. 7  | 0               | 100     | 8.6                                |

【0014】

【発明の効果】以上の説明の通り、本発明によるコーゼライト・セラミック体の製造方法はコーゼライト・セラミック体の製造過程において回収される未焼成の再生原料を用いてコーゼライト・セラミック体を製造す

る際に、厳密な粉砕条件制御が不要で、かつ再生原料の歩留まりが良好なコーゼライト・セラミック体の製造を可能にするので、コストの低減に寄与するところが大きい。

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CLAIMS

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[Claim(s)]

[Claim 1] It is the approach of manufacturing a cordierite ceramic object from the non-calcinated playback raw material which are collected in the manufacture process of a cordierite ceramic object and which consists of a predetermined cordierite-ized raw material formulation. After removing some binders [ at least ] contained in a playback raw material from a playback raw material The manufacture approach of the cordierite ceramic object characterized by producing a playback plastic matter, and fabricating and calcinating this playback plastic matter by grinding this playback raw material, producing a pulverized powder, and adding moisture, a binder, etc. at this pulverized powder, and kneading.

[Claim 2] The manufacture approach of the cordierite ceramic object indicated to claim 1 characterized by producing a playback plastic matter by adding and kneading an original start raw material and original moisture, a binder, etc. to a pulverized powder.

[Claim 3] The manufacture approach of the cordierite ceramic object indicated to claim 1 characterized by removing some binders [ at least ] contained in a playback raw material by heating from a playback raw material.

[Claim 4] The manufacture approach of the cordierite ceramic object indicated to claim 3 characterized by using methyl cellulose as a binder.

[Claim 5] The manufacture approach of the cordierite ceramic object indicated to claim 4 characterized by removing some binders [ at least ] contained in a playback raw material by heating at the temperature of 280 degrees C or more from a playback raw material.

[Claim 6] The manufacture approach of the cordierite ceramic object indicated to claim 4 characterized by removing some binders [ at least ] contained in a playback raw material by heating at 280-degree-C or more temperature of 580 degrees C or less from a playback raw material.

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## DETAILED DESCRIPTION

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### [Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacture approach of a cordierite ceramic object of manufacturing a cordierite ceramic object using the non-calcinated playback raw material collected in the manufacture process of a cordierite ceramic object.

[0002]

[Description of the Prior Art] It is known that a cordierite ceramic object has thermal resistance, and a low coefficient of thermal expansion is shown in a large temperature requirement. For this reason, the cordierite ceramic object attracts attention especially as honeycomb catalyst support for exhaust gas purification as which high thermal shock resistance is required. Usually, it calcinates, after carrying out extrusion molding of the plastic matter which mixed and kneaded shaping assistants, such as a solvent and an organic binder, and obtained them to ceramic raw materials, such as a kaolin, talc, and an alumina, through the mouthpiece for the honeycomb structures, when manufacturing a cordierite ceramic object. In order to make low the coefficient of thermal expansion of a cordierite ceramic object, in the above-mentioned production process, it is necessary to optimize the diameter of the particle of a start raw material, a raw material presentation, etc. In order to manufacture a cordierite ceramic object economically, it is desirable to carry out playback use, using as a playback raw material trash, such as a non-calcinated desiccation Plastic solid excepted in case it shifts to a baking process from a forming cycle, or its fragment.

[0003]

[Problem(s) to be Solved by the Invention] However, in order to use it as a playback raw material, when trash, such as a non-calcinated desiccation Plastic solid or its fragment, is ground, in many cases, compared with the cordierite ceramic object manufactured using the original start raw material, a coefficient of thermal expansion is large, thermal shock resistance falls, and the cordierite ceramic object manufactured using the pulverized powder has a problem of it becoming impossible to use it as honeycomb catalyst support for exhaust gas purification. Then, the method of adjusting a playback raw material is proposed so that the X diffraction peak intensity of the PUROTO enstatite side and cordierite side in the pressing side of a cordierite ceramic object may become constant value as a solution of such a problem (JP,3-72032,B). In this patent, there is a publication that it is desirable to pulverize a playback raw material after coarse grinding with a roll crusher with a gear tooth, using a pin mill as a concrete approach of grinding a playback raw material. However, a difference arises in a playback raw material, and the coefficient of thermal expansion of the acquired cordierite ceramic object needs to change a lot, and needs to control those grinding conditions by the input of the number of the pin in pin mill grinding, peripheral speed, and an ingredient strictly. moreover, the cordierite ceramic object manufactured using the original start raw material even when it ground on the optimal conditions -- comparing -- like  $0.6 \times 10^{-7} / \text{degree C}$  -- a high coefficient of thermal expansion is shown and the equivalent thing is not obtained. Moreover, the approach a strict grinding conditional control removes a pulverized powder with a particle size of less than 1mm, and uses only the pulverized powder which remained as an

unnecessary approach is proposed (JP,8-119726,A). However, as for this approach, the yield has a problem economically bad in order to remove a pulverized powder with a particle size of less than 1mm. The grinding conditional control of this invention strict in case a cordierite ceramic object is manufactured using the non-calcinated playback raw material collected in the manufacture process of a cordierite ceramic object is unnecessary, and the yield is good and it aims at offering the manufacture approach of a cordierite ceramic object which shows a coefficient of thermal expansion comparable as the case where an original start raw material is moreover used.

[0004]

[Means for Solving the Problem] This inventions are collected in the manufacture process of a cordierite ceramic object. It is the approach of manufacturing a cordierite ceramic object from the non-calcinated playback raw material which consists of a predetermined cordierite-sized raw material formulation. After removing some binders [ at least ] contained in a playback raw material from a playback raw material It is the manufacture approach of the cordierite ceramic object characterized by producing a playback plastic matter, and fabricating and calcinating this playback plastic matter by grinding this playback raw material, producing a pulverized powder, and adding moisture, a binder, etc. at this pulverized powder, and kneading. In this invention, the most important point is removing some binders [ at least ] from the above-mentioned playback raw material.

[0005]

[Function] In case a playback raw material is used, it is desirable to grind, before kneading so that uniform mixing with moisture, a binder, etc. may become easy, and to decompose into suitable magnitude. However, since the playback raw material is combined comparatively firmly with binders, such as methyl cellulose, PVA and CMC, a starch paste, and a glycerol, the comparatively big force is needed for grinding. However, the raw material particle which has stratified configurations, such as talc usually used as a cordierite-sized raw material and a kaolin, is comparatively soft, and tends to break to a mechanical shock. In JP,3-72032,B, in order to cause the reaction which completely differed from the detailed crack and the case where the mechanochemical reaction of a crystal structure was further easy to be caused, for this reason an original start raw material is used in a cordierite reaction process, to the raw material particle by the impact at the time of grinding, there is a publication that a coefficient of thermal expansion rises. Then, by removing some binders [ at least ] from a playback raw material before grinding, the bonding strength of a binder is decreased thru/or extinguished and grinding by few force is enabled. Grinding becomes possible, without deteriorating a raw material particle, without performing a strict grinding conditional control, since the impact at the time of grinding falls remarkably by grinding by few force. If all are removed, the bonding strength as a binder will be lost, grinding will also become easy, but if at least a part is removed by extent to which bonding strength becomes to some extent weak, the binder removed out of a playback raw material is enough, and does not all necessarily need to be removed. As an approach of removing a binder, although there are chemical-cleaning methods, such as the heating method and acid washing, etc., when using an organic binder, removal by the heating method is suitable also man day-wise and in facility. When a binder is methyl cellulose, since 98% or more decomposes and grinding becomes easy at 280 degrees C whenever [ stoving temperature ], whenever [ stoving temperature ] has desirable 280 degrees C or more. 99% or more decomposes [ whenever / stoving temperature ] at 580 degrees C, and more than this, since heating at high temperature is not desirable from an economical field, whenever [ stoving temperature ] has 280 degrees C or more 580 degrees C or less more desirable [ moreover, ].

[0006]

[Embodiment of the Invention] The manufacture approach of the cordierite ceramic object in the example of this invention is explained. The playback raw material to be used is a desiccation Plastic solid which is not calcinated [ of 0.17mm / in cell wall thickness of combination as shown in Table 1 /, and pitch 1.27mm, the major axis of 143mm, 98mm of minor axes, and the shape of a honeycomb with a die length of 152.4mm ]. Furnace cooling was part [ for programming-rate/of 7.5 degrees C ] carried out after the temperature up to 280 degrees C or 580 degrees C to 2kg of this Plastic solid. After cooling and a roll crusher ground the Plastic solid to the room temperature, and the pulverized powder was produced.

In order to stop grinding at this time to only break down honeycomb structure, the roll face to face dimension could be 20 slightly largermm than the sum total of the thickness of the honeycomb wall of the major-axis direction. The water 31.5 section, the methyl cellulose 3.5 section, and the stearin acid 0.5 section were added and kneaded to the pulverized-powder 100 section, and the playback plastic matter was produced. This playback plastic matter was fabricated by the extrusion method using metal mold. This Plastic solid is a honeycomb object with 0.17mm [ in cell wall thickness ], and pitch 1.27mm, a diameter [ of 30mm ], and a die length of 100mm. Next, after drying this Plastic solid using a high frequency dryer, it calcinated for 4 hours and 1400 degrees C of cordierite ceramic honeycomb-like objects were acquired.

[0007]

[Table 1]

| 成分           |              | 配 合 ( w<br>t % ) |
|--------------|--------------|------------------|
| セラミックス<br>原料 | 生カオリン        | 20.0             |
|              | 仮焼カオリン       | 10.0             |
|              | タルク          | 40.0             |
|              | アルミナ         | 15.0             |
|              | 水酸化アルミ       | 7.0              |
|              | シリカ          | 8.0              |
| 有機バインダ<br>ー  | メチルセルロ<br>ース | 3.5              |
| 潤滑剤          | ステアリン酸       | 0.5              |
| 水            |              | 31.5             |

[0008] As an example of a comparison, the cordierite ceramic object was manufactured by the following approaches. The desiccation Plastic solid of Table 1 was pulverized using the impact crusher. An impact crusher is equipment which grinds between the collision plate currently fixed and the blow plate attached in the rotor to rotate here. The pulverized powder was sorted out after grinding using various kinds of screens to five kinds, less than 0.2mm, 0.2mm or more less than 0.5mm, 0.5mm or more less than 1.0mm, 1.0mm or more less than 2.0mm, and 2.0mm or more less than 4.0mm. The water 30.3 section was added and kneaded to the pulverized-powder 100 section of each size range, and the playback plastic matter was produced. This playback plastic matter was fabricated by the extrusion method using metal mold. This Plastic solid is a honeycomb object with 0.17mm [ in cell wall thickness ], and pitch 1.27mm, a diameter [ of 30mm ], and a die length of 100mm. Next, after drying this Plastic solid using a high frequency dryer, it calcinated for 4 hours and 1400 degrees C of cordierite ceramic honeycomb-like objects were acquired. Moreover, the original start raw material was used, the plastic matter of the same combination as the desiccation Plastic solid of Table 1 was produced, it fabricated and calcinated by the same approach as the above, and the cordierite ceramic honeycomb-like object was acquired.

[0009] The average coefficient of thermal expansion in 40-800 degrees C of the cordierite ceramic object produced in the example and the example of a comparison was measured. A result is shown in Table 2. The coefficient of thermal expansion comparable as the cordierite ceramic object which used the original start raw material also in any whenever [ stoving temperature / whose ] are 280 degrees C and 580 degrees C in the example was obtained. In the pulverized powder ground by the impact crusher, in the particle size of less than 0.5mm, a coefficient of thermal expansion rises with the fall of particle size, and a coefficient of thermal expansion is almost fixed in the particle size of 0.5mm or more. However, the coefficient of thermal expansion in which particle size is also high compared with the cordierite ceramic object which used the original start raw material.

[0010]

[Table 2]

|         | N<br>o. | 再生原料の粉碎方法             | 粒度範囲 (m<br>m) | 熱膨張係数<br>( $\times 10^{-7}$<br>/℃) |
|---------|---------|-----------------------|---------------|------------------------------------|
| 実施<br>例 | 1       | 280℃に加熱後、ロールクラッシャにて粉碎 |               | 6.3                                |
|         | 2       | 580℃に加熱後、ロールクラッシャにて粉碎 |               | 6.2                                |
| 比較<br>例 | 3       | インパクトクラッシャにより粉碎       | 0.2未満         | 12.2                               |
|         | 4       |                       | 0.2以上0.5未満    | 9.6                                |
|         | 5       |                       | 0.5以上1.0未満    | 8.5                                |
|         | 6       |                       | 1.0以上2.0未満    | 8.5                                |
|         | 7       |                       | 2.0以上4.0未満    | 8.6                                |

[0011] The playback raw material of example No.1 and the original start raw material were blended at a rate shown in the following table 3, the water 31.5 section, the methyl cellulose 3.5 section, and the stearin acid 0.5 section were further added and kneaded to this compound 100 section, and the playback plastic matter was produced. Moreover, the playback raw material of example No. of comparison 7 in an example 1 and the original start raw material were blended at a rate shown in the following table 3, the water of optimum dose, methyl cellulose, and stearin acid were added and kneaded, and the playback plastic matter was produced. These playback plastic matters were fabricated by the extrusion method using metal mold. This Plastic solid is a honeycomb object with 0.17mm [ in cell wall thickness ], and pitch 1.27mm, a diameter [ of 30mm ], and a die length of 100mm. Next, after drying this Plastic solid using a high frequency dryer, it calcinated for 4 hours and 1400 degrees C of cordierite ceramic honeycomb-like objects were acquired.

[0012] The average coefficient of thermal expansion in 40-800 degrees C of the cordierite ceramic object produced above was measured. A result is collectively shown in Table 3. In the example, no matter it might blend at what rate, the coefficient of thermal expansion comparable as the cordierite ceramic object which used the original start raw material was obtained. On the other hand, in the example of a comparison, although a coefficient of thermal expansion comparable as the cordierite ceramic object which used the start raw material with 10 original% of rates of a compounding ratio is obtained, if it blends more than it, the rise of a coefficient of thermal expansion will be accepted.

[0013]

[Table 3]

| N<br>o. | 使用再生原料 | 配合比率    |      | 熱膨張係数<br>( $\times 10^{-7}$<br>/℃) |
|---------|--------|---------|------|------------------------------------|
|         |        | 元来の出発原料 | 再生原料 |                                    |
| 8       | —      | 100     | 0    | 6.2                                |
| 9       | N o. 1 | 80      | 20   | 6.2                                |
| 10      | N o. 1 | 60      | 40   | 6.3                                |
| 11      | N o. 1 | 40      | 60   | 6.2                                |
| 12      | N o. 1 | 20      | 80   | 6.2                                |
| 1       | N o. 1 | 0       | 100  | 6.3                                |
| 13      | N o. 7 | 95      | 5    | 6.3                                |
| 14      | N o. 7 | 90      | 10   | 6.2                                |
| 15      | N o. 7 | 80      | 20   | 7.4                                |
| 16      | N o. 7 | 60      | 40   | 8.7                                |
| 7       | N o. 7 | 0       | 100  | 8.6                                |

[0014]

[Effect of the Invention] Since a strict grinding conditional control makes unnecessary manufacture of a cordierite ceramic object with the good yield of a playback raw material possible in case the manufacture approach of the cordierite ceramic object according to this invention as the above explanation manufactures a cordierite ceramic object using the non-calcinated playback raw material collected in the manufacture process of a cordierite ceramic object, the place which contributes to

reduction of cost is large.

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[Translation done.]

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TECHNICAL FIELD

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[Field of the Invention] This invention relates to the manufacture approach of a cordierite ceramic object of manufacturing a cordierite ceramic object using the non-calcinated playback raw material collected in the manufacture process of a cordierite ceramic object.

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PRIOR ART

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[Description of the Prior Art] It is known that a cordierite ceramic object has thermal resistance, and a low coefficient of thermal expansion is shown in a large temperature requirement. For this reason, the cordierite ceramic object attracts attention especially as honeycomb catalyst support for exhaust gas purification as which high thermal shock resistance is required. Usually, it calcinates, after carrying out extrusion molding of the plastic matter which mixed and kneaded shaping assistants, such as a solvent and an organic binder, and obtained them to ceramic raw materials, such as a kaolin, talc, and an alumina, through the mouthpiece for the honeycomb structures, when manufacturing a cordierite ceramic object. In order to make low the coefficient of thermal expansion of a cordierite ceramic object, in the above-mentioned production process, it is necessary to optimize the diameter of the particle of a start raw material, a raw material presentation, etc. In order to manufacture a cordierite ceramic object economically, it is desirable to carry out playback use, using as a playback raw material trash, such as a non-calcinated desiccation Plastic solid excepted in case it shifts to a baking process from a forming cycle, or its fragment.

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EFFECT OF THE INVENTION

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[Effect of the Invention] Since a strict grinding conditional control makes unnecessary manufacture of a cordierite ceramic object with the good yield of a playback raw material possible in case the manufacture approach of the cordierite ceramic object according to this invention as the above explanation manufactures a cordierite ceramic object using the non-calcinated playback raw material collected in the manufacture process of a cordierite ceramic object, the place which contributes to reduction of cost is large.

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**TECHNICAL PROBLEM**

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[Problem(s) to be Solved by the Invention] However, in order to use it as a playback raw material, when trash, such as a non-calcinated desiccation Plastic solid or its fragment, is ground, in many cases, compared with the cordierite ceramic object manufactured using the original start raw material, a coefficient of thermal expansion is large, thermal shock resistance falls, and the cordierite ceramic object manufactured using the pulverized powder has a problem of it becoming impossible to use it as honeycomb catalyst support for exhaust gas purification. Then, the method of adjusting a playback raw material is proposed so that the X diffraction peak intensity of the PUROTO enstatite side and cordierite side in the pressing side of a cordierite ceramic object may become constant value as a solution of such a problem (JP,3-72032,B). In this patent, there is a publication that it is desirable to pulverize a playback raw material after coarse grinding with a roll crusher with a gear tooth, using a pin mill as a concrete approach of grinding a playback raw material. However, a difference arises in a playback raw material, and the coefficient of thermal expansion of the acquired cordierite ceramic object needs to change a lot, and needs to control those grinding conditions by the input of the number of the pin in pin mill grinding, peripheral speed, and an ingredient strictly. moreover, the cordierite ceramic object manufactured using the original start raw material even when it ground on the optimal conditions -- comparing -- like  $0.6 \times 10^{-7} / \text{degree C}$  -- a high coefficient of thermal expansion is shown and the equivalent thing is not obtained. Moreover, the approach a strict grinding conditional control removes a pulverized powder with a particle size of less than 1mm, and uses only the pulverized powder which remained as an unnecessary approach is proposed (JP,8-119726,A). However, as for this approach, the yield has a problem economically bad in order to remove a pulverized powder with a particle size of less than 1mm. The grinding conditional control of this invention strict in case a cordierite ceramic object is manufactured using the non-calcinated playback raw material collected in the manufacture process of a cordierite ceramic object is unnecessary, and the yield is good and it aims at offering the manufacture approach of a cordierite ceramic object which shows a coefficient of thermal expansion comparable as the case where an original start raw material is moreover used.

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MEANS

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[Means for Solving the Problem] This inventions are collected in the manufacture process of a cordierite ceramic object. It is the approach of manufacturing a cordierite ceramic object from the non-calcinated playback raw material which consists of a predetermined cordierite-ized raw material formulation. After removing some binders [ at least ] contained in a playback raw material from a playback raw material It is the manufacture approach of the cordierite ceramic object characterized by producing a playback plastic matter, and fabricating and calcinating this playback plastic matter by grinding this playback raw material, producing a pulverized powder, and adding moisture, a binder, etc. at this pulverized powder, and kneading. In this invention, the most important point is removing some binders [ at least ] from the above-mentioned playback raw material.

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## OPERATION

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[Function] In case a playback raw material is used, it is desirable to grind, before kneading so that uniform mixing with moisture, a binder, etc. may become easy, and to decompose into suitable magnitude. However, since the playback raw material is combined comparatively firmly with binders, such as methyl cellulose, PVA and CMC, a starch paste, and a glycerol, the comparatively big force is needed for grinding. However, the raw material particle which has stratified configurations, such as talc usually used as a cordierite-ized raw material and a kaolin, is comparatively soft, and tends to break to a mechanical shock. In JP,3-72032,B, in order to cause the reaction which completely differed from the detailed crack and the case where the mechanochemical reaction of a crystal structure was further easy to be caused, for this reason an original start raw material is used in a cordierite reaction process, to the raw material particle by the impact at the time of grinding, there is a publication that a coefficient of thermal expansion rises. Then, by removing some binders [ at least ] from a playback raw material before grinding, the bonding strength of a binder is decreased thru/or extinguished and grinding by few force is enabled. Grinding becomes possible, without deteriorating a raw material particle, without performing a strict grinding conditional control, since the impact at the time of grinding falls remarkably by grinding by few force. If all are removed, the bonding strength as a binder will be lost, grinding will also become easy, but if at least a part is removed by extent to which bonding strength becomes to some extent weak, the binder removed out of a playback raw material is enough, and does not all necessarily need to be removed. As an approach of removing a binder, although there are chemical-cleaning methods, such as the heating method and acid washing, etc., when using an organic binder, removal by the heating method is suitable also man day-wise and in facility. When a binder is methyl cellulose, since 98% or more decomposes and grinding becomes easy at 280 degrees C whenever [ stoving temperature ], whenever [ stoving temperature ] has desirable 280 degrees C or more. 99% or more decomposes [ whenever / stoving temperature ] at 580 degrees C, and more than this, since heating at high temperature is not desirable from an economical field, whenever [ stoving temperature ] has 280 degrees C or more 580 degrees C or less more desirable [ moreover, ].

[0006]

[Embodiment of the Invention] The manufacture approach of the cordierite ceramic object in the example of this invention is explained. The playback raw material to be used is a desiccation Plastic solid which is not calcinated [ of 0.17mm / in cell wall thickness of combination as shown in Table 1 /, and pitch 1.27mm, the major axis of 143mm, 98mm of minor axes, and the shape of a honeycomb with a die length of 152.4mm ]. Furnace cooling was part [ for programming-rate/of 7.5 degrees C ] carried out after the temperature up to 280 degrees C or 580 degrees C to 2kg of this Plastic solid. After cooling and a roll crusher ground the Plastic solid to the room temperature, and the pulverized powder was produced. In order to stop grinding at this time to only break down honeycomb structure, the roll face to face dimension could be 20 slightly largermmm than the sum total of the thickness of the honeycomb wall of the major-axis direction. The water 31.5 section, the methyl cellulose 3.5 section, and the stearin acid 0.5 section were added and kneaded to the pulverized-powder 100 section, and the playback plastic matter was produced. This playback plastic matter was fabricated by the extrusion method using metal

mold. This Plastic solid is a honeycomb object with 0.17mm [ in cell wall thickness ], and pitch 1.27mm, a diameter [ of 30mm ], and a die length of 100mm. Next, after drying this Plastic solid using a high frequency dryer, it calcinated for 4 hours and 1400 degrees C of cordierite ceramic honeycomb-like objects were acquired.

[0007]

[Table 1]

| 成分           |          | 配 合 ( w<br>t % ) |
|--------------|----------|------------------|
| セラミックス<br>原料 | 生カオリン    | 20.0             |
|              | 仮焼カオリン   | 10.0             |
|              | タルク      | 40.0             |
|              | アルミナ     | 15.0             |
|              | 水酸化アルミ   | 7.0              |
|              | シリカ      | 8.0              |
| 有機バインダー      | メチルセルロース | 3.5              |
| 潤滑剤          | ステアリン酸   | 0.5              |
| 水            |          | 31.5             |

[0008] As an example of a comparison, the cordierite ceramic object was manufactured by the following approaches. The desiccation Plastic solid of Table 1 was pulverized using the impact crusher. An impact crusher is equipment which grinds between the collision plate currently fixed and the blow plate attached in the rotor to rotate here. The pulverized powder was sorted out after grinding using various kinds of screens to five kinds, less than 0.2mm, 0.2mm or more less than 0.5mm, 0.5mm or more less than 1.0mm, 1.0mm or more less than 2.0mm, and 2.0mm or more less than 4.0mm. The water 30.3 section was added and kneaded to the pulverized-powder 100 section of each size range, and the playback plastic matter was produced. This playback plastic matter was fabricated by the extrusion method using metal mold. This Plastic solid is a honeycomb object with 0.17mm [ in cell wall thickness ], and pitch 1.27mm, a diameter [ of 30mm ], and a die length of 100mm. Next, after drying this Plastic solid using a high frequency dryer, it calcinated for 4 hours and 1400 degrees C of cordierite ceramic honeycomb-like objects were acquired. Moreover, the original start raw material was used, the plastic matter of the same combination as the desiccation Plastic solid of Table 1 was produced, it fabricated and calcinated by the same approach as the above, and the cordierite ceramic honeycomb-like object was acquired.

[0009] The average coefficient of thermal expansion in 40-800 degrees C of the cordierite ceramic object produced in the example and the example of a comparison was measured. A result is shown in Table 2. The coefficient of thermal expansion comparable as the cordierite ceramic object which used the original start raw material also in any whenever [ stoving temperature / whose ] are 280 degrees C and 580 degrees C in the example was obtained. In the pulverized powder ground by the impact crusher, in the particle size of less than 0.5mm, a coefficient of thermal expansion rises with the fall of particle size, and a coefficient of thermal expansion is almost fixed in the particle size of 0.5mm or more. However, the coefficient of thermal expansion in which particle size is also high compared with the cordierite ceramic object which used the original start raw material.

[0010]

[Table 2]

|          | N<br>o. | 再生原料の粉碎方法                 | 粒 径 範 囲 (m<br>m) | 熱膨張係数<br>( $\times 10^{-7}$<br>/℃) |
|----------|---------|---------------------------|------------------|------------------------------------|
| 実 施<br>例 | 1       | 280℃に加熱後、ロールク<br>ラッシャにて粉碎 |                  | 6.3                                |
|          | 2       | 580℃に加熱後、ロールク<br>ラッシャにて粉碎 |                  | 6.2                                |
| 比 較<br>例 | 3       | インパクトクラッシャ<br>により粉碎       | 0.2未満            | 12.2                               |
|          | 4       |                           | 0.2以上0.5<br>未満   | 9.5                                |
|          | 5       |                           | 0.5以上1.0<br>未満   | 8.5                                |
|          | 6       |                           | 1.0以上2.0<br>未満   | 8.5                                |
|          | 7       |                           | 2.0以上4.0<br>未満   | 8.6                                |

[0011] The playback raw material of example No.1 and the original start raw material were blended at a rate shown in the following table 3, the water 31.5 section, the methyl cellulose 3.5 section, and the stearin acid 0.5 section were further added and kneaded to this compound 100 section, and the playback plastic matter was produced. Moreover, the playback raw material of example No. of comparison 7 in an example 1 and the original start raw material were blended at a rate shown in the following table 3, the water of optimum dose, methyl cellulose, and stearin acid were added and kneaded, and the playback plastic matter was produced. These playback plastic matters were fabricated by the extrusion method using metal mold. This Plastic solid is a honeycomb object with 0.17mm [ in cell wall thickness ], and pitch 1.27mm, a diameter [ of 30mm ], and a die length of 100mm. Next, after drying this Plastic solid using a high frequency dryer, it calcinated for 4 hours and 1400 degrees C of cordierite ceramic honeycomb-like objects were acquired.

[0012] The average coefficient of thermal expansion in 40-800 degrees C of the cordierite ceramic object produced above was measured. A result is collectively shown in Table 3. In the example, no matter it might blend at what rate, the coefficient of thermal expansion comparable as the cordierite ceramic object which used the original start raw material was obtained. On the other hand, in the example of a comparison, although a coefficient of thermal expansion comparable as the cordierite ceramic object which used the start raw material with 10 original% of rates of a compounding ratio is obtained, if it blends more than it, the rise of a coefficient of thermal expansion will be accepted.

[0013]

[Table 3]

| N<br>o. | 使用再生原<br>料 | 配合比率            |            | 熱膨張係数<br>( $\times 10^{-7}$<br>/℃) |
|---------|------------|-----------------|------------|------------------------------------|
|         |            | 元 来 の 出 発<br>原料 | 再 生 原<br>料 |                                    |
| 8       | —          | 100             | 0          | 6.2                                |
| 9       | No. 1      | 80              | 20         | 6.2                                |
| 10      | No. 1      | 60              | 40         | 6.3                                |
| 11      | No. 1      | 40              | 60         | 6.2                                |
| 12      | No. 1      | 20              | 80         | 6.2                                |
| 1       | No. 1      | 0               | 100        | 6.3                                |
| 13      | No. 7      | 95              | 5          | 6.3                                |
| 14      | No. 7      | 90              | 10         | 6.2                                |
| 15      | No. 7      | 80              | 20         | 7.4                                |
| 16      | No. 7      | 60              | 40         | 8.7                                |
| 7       | No. 7      | 0               | 100        | 8.6                                |

[Translation done.]

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